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The Effect of the Induced Uniaxial Anisotropy on the Domain Wall Displacements and Magnetic Behavior of Ferromagnetic Cubic Solid Solutions*

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Synopsis

When a ferromagnetic cubic solid solution is heat-treated in the absence of external magnetic field, the uniaxial anisotropy is induced along magnetization vectors oriented in compliance with the domain distribution present during the heat-treatment. This induced uniaxial anisotropy affects the displacement of a domain wall, or stabilizes it, since the rotation of a magnetization vector within the domain wall from its original direction results an increase in the induced uniaxial anisotropy energy. The restoring force acting on the domain wall as a function of the wall displacement has been calculated for cubic solid solutions with various crystal lattices. The results of calculation lead us to the conclusions that the permeability in low fields may be reduced appreciably by the effect of domain wall stabilization and the peculiar magnetic properties such as exhibited by perminvars are quite common to annealed or slowly cooled cubic solid solutions in which the uniaxial anisotropy can be induced along the directions of easy magnetization. Further, the influence of heat-treatment on the magnetic properties of cubic solid solutions is discussed in terms of the stabilization of domain walls by the induced uniaxial anisotropy and a reference is also made to the permalloy problem.

I. Introduction

Previously, Néel[(1)] proposed, in a paper on a theory of magnetic aftereffect due to the diffusion of interstitial atoms in a ferromagnetic metal, such as carbon or nitrogen atoms in α-iron, the exsistence of a magnetocrystalline coupling between the arrangement of interstitial atoms and the direction of magnetization vectors. This coupling induces a time-dependent uniaxial anisotropy and accordingly a time-dependent restoring force acting on domain walls, resulting the magnetic aftereffect. As has recently been indicated by the present author and Yamamoto[(2)] and by Néel[(3)], independently of each other, in the theory of the uniaxial ferromagnetic anisotropy induced by magnetic annealing, there exists, in a ferromagnetic solid solution of the substitutional type, a similar magnetocrystalline coupling between the arrangements of solute atom pairs and the direction of

* The 834th report of the Research Institute for Iron, Steel and Other Metals. A preliminary report written in Japanese was published in Nippon Kinzoku Gakkai-shi, **19** (1955), 127 in collaboration with M. Yamamoto.

(1) L. Néel, J. de phys., **12** (1951), 339; **13** (1952), 249.

(2) S. Taniguchi and M. Yamamoto, Sci. Rep. RITU, **A6** (1954), 330; S. Taniguchi, Sci. Rep. RITU, **A7** (1955), 269.

(3) L. Néel, J. de phys., **15** (1954), 225.

magnetization, which may induce a uniaxial anisotropy acting *surely* as a restoring force on domain walls. In a substitutional solid solution, the solubility limit is relatively high, and hence the displacement of domain walls will be affected by the induced uniaxial anisotropy far more conspicuously than in an interstitial solid solution.

In this paper, we calculate, for cubic solid solutions of the substitutional type, the restoring force due to the induced uniaxial anisotropy acting on a domain wall as a function of its displacement, using the results obtained in previous papers(2), and show that the peculiar magnetic properties, such as revealed by the so-called "perminvar"(4), are really common to annealed or slowly cooled cubic solid solutions of the substitutional type in which the uniaxial anisotropy can be induced along the directions of easy magnetization. Further, brief discussions are given on the general aspect of the influence of heat-treatment upon the magnetic properties of cubic solid solutions and on the permalloy problem.

II. Uniaxial anisotropy induced by magnetic annealing

When a ferromagnetic cubic solid solution of the substitutional type is heat-treated in the presence of magnetic field, a uniaxial ferromagnetic anisotropy is induced, superposing the proper cubic magnetocrystalline anisotropy. The origin of this induced uniaxial anisotropy has been interpreted by us(2) as follows:- Among atoms in a ferromagnetic crystal, there exists, besides the ordinary exchange interaction, an anisotropic interaction due to the interplay between the spin-orbit interaction and orbital valence, which is thought to be the origin of the magnetocrystalline anisotropy and can be expressed approximately in the form of dipole-dipole and quadrupole-quadrupole couplings(5). In a ferromagnetic solid solution, this anisotropic interaction may generally differ for different kinds of atom pairs such as A-A, A-B and B-B atom pairs in solid solution of atoms A and B. Accordingly, the equilibrium distribution of solute atom pairs becomes anisotropic below the Curie temperature, only if the diffusion of atoms can take place at that temperature, and it depends on the direction of magnetization vector in regard to the crystallographic axes. This anisotropic distribution of solute atom pairs may yield a uniaxial anisotropy of magnetocrystalline origin in cubic solid solution.

If the distribution of solute atom pairs does not change after it attained the equilibrium state at a temperature Θ, the uniaxial anisotropy, F, at a temperature T is given approximately by

$$F = -K_u \Omega \,, \tag{1}$$

where

$$K_u = A_1 N n^2 C^2 B_T{}^2 B_\Theta{}^2 / k\Theta \tag{2}$$

and

$$\Omega = A_2 \textstyle\sum_i \alpha_i^2 \beta_i^2 + A_3 \sum_{j>i} \alpha_i \alpha_j \beta_i \beta_j + A_4 \,. \tag{3}$$

(4) See, for example, R. M. Bozorth, *Ferromagnetism*, New York (1951), p. 160.
(5) J. H. Van Vleck, Phys. Rev., **52** (1937), 1178.

Here, α_i's and β_i's $(i = 1, 2, 3)$ are the direction cosines, referred to the crystal axes, of magnetization vectors at temperatures T and Θ, respectively, and N the number of atoms per unit volume, n the concentration of solute atoms, and k the Boltzman constant. Further, $C = C_{AA}+C_{BB}-2C_{AB}$, in which C_{AA}, C_{AB} and C_{BB} are dipole-dipole coupling constants of atom pairs A–A, A–B and B–B, respectively, and B_T and B_Θ are the averaged magnetizations at temperatures T and Θ, respectively. A_1 in Eq. (2) and A_2, A_3 and A_4 in Eq. (3) are numerical factors depending on the type of crystal lattice. For convenience in the following calculations, we choose A_4 so as to make the uniaxial anisotropy energy induced along one of the directions of easy magnetization as zero, and so it depends further on the sign of the cubic anisotropy constant K_c. The values of A_1(6), A_2, A_3 and A_4 are given in Table 1, which shows that, irrespective of the sign of K_c,

$$\Omega_{f.c.c.} = \Omega_{s.c.} + 4\Omega_{b.c.c.}\,. \tag{4}$$

Table 1. Values of A_1, A_2, A_3 and A_4 for various crystal lattices.

Crystal lattices	A_1	A_2	A_3	A_4	
				$K_c>0$	$K_c<0$
f. c. c.	9/2	1	4	−1	−5/3
b. c. c.	16	0	1	0	−1/3
s. c.	9	1	0	−1	−1/3

Finally, we shall refer briefly to the interstitial solid solutions. For interstitial solid solution with body-centered cubic lattice, the induced uniaxial anisotropy can be obtained only by substituting n^2 by n' (=the ratio of the number of interstitial atoms to that of solvent atoms) in the formula for simple cubic solid solution of the substitutional type. For interstitial solid solution with face-centered cubic lattice, the uniaxial anisotropy can not appear in the nearest neighbor assumption employed in our theory, if interstitial atoms occupy the body centered positions in face-centered cubic lattice, as it really seems so.

III. The restoring force acting on the domain walls stabilized by the uniaxial anisotropy and its effect upon the wall displacements

When a ferromagnetic crystal is cooled down beyond the Curie temperature in the absence of external magnetic field, it is generally divided into magnetic domains in order to minimize the magnetostatic energy due to the appearence of magnetic free poles on its surface. If the internal stress is small, the magnetization vector in a domain is oriented along one of the directions of easy magnetization determined by the magnetocrystalline anisotropy energy. Between two domains, there

(6) The values of A_1 given in Table 1 are those in the case where the dipole-dipole coupling energy is expressed as $CB_T^2(1-3\cos^2\psi)$, in which ψ is the angle between the magnetization vector and the direction of dipole pair.

exists a transition layer, named a domain wall, within which the magnetization vector rotates continuously from one domain to another.

Let us consider the displacement of domain walls in an infinite crystal of ferromagnetic cubic solid solution of the substitutional type, in which the uniaxial anisotropy is induced along the magnetization vector at temperatures below the Curie temperature, as described in Section II. We take the normal of the domain wall as the X axis and denote the position of the central layer of the wall as u. We take, further, the direction of the projection of the magnetization vector at $X-u=\infty$ on the plane of the wall as the Y axis. Then, the direction of magnetization vector, everywhere in the crystal, is specified by the two angles, θ and ϕ, where θ is an angle between the magnetization vector and X axis and ϕ an angle between the projection of the magnetization vector on the plane of the wall and Y axis. θ is contant, since the normal component of the magnetization vector to the plane of the wall must be constant, while ϕ is a function of $X-u$ or

$$\phi = G(X-u)\,. \tag{5}$$

If the crystal has been maintained at a temperature Θ for a time long enough to induce the uniaxial anisotropy and then the domain wall is displaced by an externally applied field or stress from u to u' and accordingly the angle between the Y axis and the projection of the magnetization vector at X is changed from ϕ to ϕ', ϕ' is given by the same function as Eq. (5) :-

$$\phi' = G(X-u')\,. \tag{6}$$

Here we have assumed that the shape of the domain wall never changes as it displaces, as may be allowed if the induced uniaxial anisotropy is far smaller than the cubic magnetocrystalline anisotropy. If this displacement of the wall is so rapid that the distribution of atoms may not vary, the increase in energy, F', caused by the rotation of the magnetization vector from the $(\theta,\ \phi)$ to the $(\theta,\ \phi')$ direction is given, from Eq. (1), by

$$F' = -K_u'\Omega(X-u,\ X-u')\,, \tag{7}$$

where

$$K_u' = A_1 N n^2 C^2 B_\Theta^4/k\Theta\,, \tag{8}$$

and Ω is a function of $X-u$ and $X-u'$ as obtained by replacing α_i's and β_i's in Eq. (3) by θ, ϕ, and ϕ', and using the relations (5) and (6). Integrating Eq. (7) from $X=-\infty$ to ∞, we obtain the energy, W, associated with the wall displacement $U=u'-u$ as

$$W = -K_u'S(U)\,, \tag{9}$$

where

$$S(U) = \int_{-\infty}^{\infty}\Omega(X-u,\ X-u')dX\,. \tag{10}$$

Then, the force P acting on the wall is given by

$$P = -\partial W/\partial U = K_u'\partial S(U)/\partial U = K_u'S'(U)\,. \tag{11}$$

It is to be noted that, from the relation (4),

$$S_{f.c.c.} = S_{s.c.} + 4S_{b.c.c.}, \tag{12}$$

and accordingly

$$S'_{f.c.c.} = S'_{s.c.} + 4S'_{b.c.c.}. \tag{13}$$

Now we calculate the value of $S(U)$ for some type of domain wall in various cubic lattices. As an example, we take a face-centered cubic solid solution with a positive cubic anisotropy constant ($K_c > 0$), for which Ω (Eq. (3)) is written as (cf. Table 1)

$$\Omega = \sum_i \alpha_i^2 \beta_i^2 + 4\sum_{j>i} \alpha_i \alpha_j \beta_i \beta_j - 1. \tag{14}$$

In the first place, let us consider a 90° Wall parallel to the (100) plane. If we take the [010] direction as the Y axis (the X axis is the [100] direction), the relations between ϕ and $X-u$ and between ϕ' and $X-u'$(7) are expressed, respectively, by

$$\cot\phi = \exp\{(X-u)/d_0\} \quad \text{and} \quad \cot\phi' = \exp\{(X-u')/d_0\}. \tag{15}$$

Here a measure of the domain wall width, d_0, is given by

$$d_0 = a(E/|K_c|)^{1/2}, \tag{16}$$

where a is the lattice parameter and E the exchange energy density. Since $\alpha_1 = \beta_1 = 0$, $\alpha_2 = \cos\phi$, $\alpha_3 = \sin\phi$, $\beta_2 = \cos\phi'$ and $\beta_3 = \sin\phi'$, Eq. (14) becomes(8)

$$\Omega = 2\cos^2\phi\cos^2\phi' - \cos^2\phi - \cos^2\phi' + 4\cos\phi\sin\phi\cos\phi'\sin\phi', \tag{17}$$

which reduces to

$$\begin{aligned} \Omega = & [\{a + a' - 4(aa')^{1/2}\}/(a - a')] \\ & \times [1/\{1 + a\exp(2X/d_0)\} - 1/\{1 + a'\exp(2X/d_0)\}], \end{aligned} \tag{18}$$

where $a = \exp(-2u/d_0)$ and $a' = \exp(-2u'/d_0)$. Then, integrating Eq. (18) from $X = -\infty$ to ∞, we have

$$S(U) = -U\{\coth(U/d_0) - 2\mathrm{cosech}(U/d_0)\}. \tag{19}$$

Secondly, we consider a 180° wall parallel to the (100) plane, which is displaced from $U/2$ to $-U/2$. Then, the relations between ϕ and $X - U/2$ and between ϕ' and $X+U/2$ are, respectively,

$$\left.\begin{aligned} &\cot\phi = -g\sinh\{\alpha(2X-U)/d_0\} \\ \text{and} \quad &\cot\phi' = -g\sinh\{\alpha(2X+U)/d_0\}, \end{aligned}\right\} \tag{20}$$

where

$$g = \{\tau/(1+\tau)\}^{1/2} \quad \text{and} \quad \alpha = (1+\tau)^{1/2}, \tag{21}$$

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(7) The relations between ϕ and $X-u$ for all kinds of domain walls in cubic ferromagnetic crystal with $K_c \gtrless 0$ have been given by B. A. Lilley, Phil. Mag., **41** (1950), 729.

(8) If the value of A_4 in Eq. (3) is not determined in such a way as mentioned in Section II, a constant term may appear in Eq. (17). Ignorance of this term as a standard of the energy concerned often leads to a faulted result.

and

$$\tau = 9(c_{11} - c_{12})\lambda_{100}^2/4K_c . \tag{22}$$

Here c_{11} and c_{12} are the elastic constants and λ_{100} the saturation magnetostriction along the [100] direction. The expression for Ω is the same as Eq. (17), which may also be written as

$$\Omega = 2\sin^2\phi\sin^2\phi' - \sin^2\phi - \sin^2\phi' + 4\cos\phi\sin\phi\cos\phi'\sin\phi' . \tag{17a}$$

Then, we integrate Eq. (17a) from $X = -\infty$ to ∞ to obtain the value of $S(U)$. It is to be noted, here, that the terms $\sin^2\phi$ and $\sin^2\phi'$ in Eq.(17a) give integrals independent of U. Because, if we put

$$z = \exp(2\alpha X/d_0), \quad a = \exp(\alpha U/d_0), \quad \text{and} \quad A = 1-2/g^2, \tag{23}$$

then we get

$$\sin^2\phi = 2(A + 1)az/(z^2 + 2Aaz + a^2)$$

and accordingly

$$\int_{-\infty}^{\infty} \sin^2\phi dX = (d_0/\alpha)(A+1)a\int_0^{\infty}(z^2+2Aaz+a^2)^{-1}dz$$
$$= -(d_0/2\alpha)\{(A+1)/(A-1)\}^{1/2}\log[\{A-(A^2-1)^{1/2}\}/\{A+(A^2-1)^{1/2}\}] .$$

Similarly, we obtain the same value for $\int_{-\infty}^{\infty}\sin^2\phi' dX$. Thus, we need not concern these terms. Now, we have

$$\int_{-\infty}^{\infty} 2\sin^2\phi\sin^2\phi' dX = 2(d_0/\alpha)(1+A)^2P$$
$$\times\int_0^{\infty}\left\{\frac{(a^{-1}+a)z+2a^2A}{z^2+2Aaz+a^2} - \frac{a^2(a^{-1}+a)z+2A}{a^2z^2+2Aaz+1}\right\}dz ,$$

and

$$\int_{-\infty}^{\infty} \cos\phi\sin\phi\cos\phi'\sin\phi' dX = -(d_0/\alpha)(1+A)P$$
$$\times\int_0^{\infty}\left[\frac{\{(a^{-1}+a)^2+4A\}z+a\{4A^2+2aA(a^{-1}+a)-(a^{-1}-a)(a^{-1}+a)\}}{z^2+2Aaz+a^2}\right.$$
$$\left. - \frac{a^2\{(a^{-1}+a)^2+4A\}z+a\{4A^2+2a^{-1}A(a^{-1}+a)+(a^{-1}-a)(a^{-1}+a)\}}{a^2z^2+2Aaz+1}\right]dz ,$$

where $P = [(a^{-1}-a)\{(a^{-1}+a)^2-4A^2\}]^{-1}$, and finally we get

$$\begin{aligned} S(U) = {} & [2(d_0/\alpha)(1 + \cosh V)^2\{(\alpha U/d_0)\coth(\alpha U/d_0) - V\coth V\} \\ & +4(d_0/\alpha)(1 + \cosh V)^2 V \operatorname{cosech} V \cosh(\alpha U/d_0) \\ & -4(1+\cosh V)U\{\cosh V + \cosh^2(\alpha U/d_0)\}\operatorname{cosech}(\alpha U/d_0)] \\ & \div \{\cosh^2(\alpha U/d_0) - \cosh^2 V\} , \end{aligned} \tag{24}$$

where

$$\cosh V = A = (2 + \tau)/\tau . \tag{25}$$

In the same manner, we can obtain values of $S(U)$ for other domain walls in various cubic lattices. It is to be noted, however, that, for $K_c<0$, $g=\{(1+\tau)/(8+\tau)\}^{1/2}$, $\alpha=(1/2)\{(8+\tau)/3\}^{1/2}$ and $\tau = 54c_{44}\lambda_{111}^2/|K_c|$, where c_{44} is the elastic constant and λ_{111} the saturation magnetostriction along the [111] direction. The values of $S(U)$ thus obtained are summarized in Table 2, in which only those for simple and body-centered cubic lattices are shown, since the relations (12) and (13) hold.

Table 2. Values of $S(U)$ for some kinds of domain walls in simple cubic and body-centered cubic lattices. $S(U)_{f.c.c.}=S(U)_{s.c.}+4S(U)_{b.c.c.}$.

Sign of cubic anisotropy	Kind of domain wall	$S(U)_{s.c.}$	$S(U)_{b.c.c.}$
$K_c>0$	(100) 90° wall	(a)	(b)
	(100) 180° wall	(c)	(d)
	(110) 180° wall	(c)	(1/8)(c)+(d)
$K_c<0$	(100) 71° wall	(e)	(f)
	(110) 71° wall	(g)	(h)
	(112) 180° wall	(1/12)(c)+(4/3)(d)	(33/72)(c)+(1/3)(d)

(a) $= -U\coth(U/d_0)$,
(b) $= (U/2)\,\mathrm{cosech}(U/d_0)$,
(c) $= 2(d_0/\alpha)(1+\cosh V)^2\{(\alpha U/d_0)\coth(\alpha U/d_0)-V\coth V\}/\{\cosh^2(\alpha U/d_0)-\cosh^2 V\}$,
(d) $= [(d_0/\alpha)(1+\cosh V)^2\, V\,\mathrm{cosech}\, V\cosh(\alpha U/d_0)$
$\quad -(1+\cosh V)U\{\cosh V+\cosh^2(\alpha U/d_0)\}\mathrm{cosech}(\alpha U/d_0)]/\{\cosh^2(\alpha U/d_0)-\cosh^2 V\}$,
(e) $= (4/9)U\,\mathrm{cosech}(\sqrt{2/3}\,U/d_0)$,
(f) $= (2/9)[-U\coth(\sqrt{2/3}\,U/d_0)+\sqrt{3/2}\,d_0\log 2-\sqrt{3/2}\,d_0\log\{1+\cosh(\sqrt{2/3}\,U/d_0\}]$,
(g) $= (4/3)[-2U\coth(U/2d_0)+\sqrt{2}\,d_0\{(\pi/2)-\tan^{-1}(1/2\sqrt{2})\}]/\{1-9\cosh^2(U/2d_0)\}$,
(h) $= [1-(25/6)/\{1-9\cosh^2(U/2d_0)\}][-U\coth(U/2d_0)+(1/\sqrt{2})d_0\{(\pi/2)-\tan^{-1}(1/2\sqrt{2})\}]$.

Figs. 1~6 show $S'(U)$ as functions of U/d_0 for some domain walls, which were calculated using the following values of material constants :- $|\lambda_{100}|=|\lambda_{111}|=10\times10^{-6}$, $c_{11}-c_{12}=c_{44}=1.0\times10^{12}$ dyne/cm² and $|K_c|=1.0\times10^4$ erg/cm³. As seen from these figures, $S'(U)$'s for non-180° walls are negative throughout and those for 180° walls are also negative for almost all parts of the curves, indicating, as may be expected, that the force, P, acting on a domain wall is a restoring force or the wall is stabilized. Further, the curves for non-180° walls (Figs. 1~3) may be divided into two groups. In the first group, $S'(U)$ is relatively low and approaches to zero as U increases, while in the second group $S'(U)$ remains finite. The body-centered cubic solid solution with a positive K_c and simple cubic solid solution with a negative K_c belong to the first group and the remaining solid solutions to the second group. The curves for 180° walls (Figs. 4~6), however, resemble to each other in that $-S'(U)$, after passing over a maximum and, further by circumstances, a small minimum, approaches to zero as U increases. It is to be noted that, in solid solutions belonging to the first group of non-180° wall curves, the uniaxial anisotropy can not be induced along the directions of easy magnetization(2), showing a slight stabilization only within the wall, and

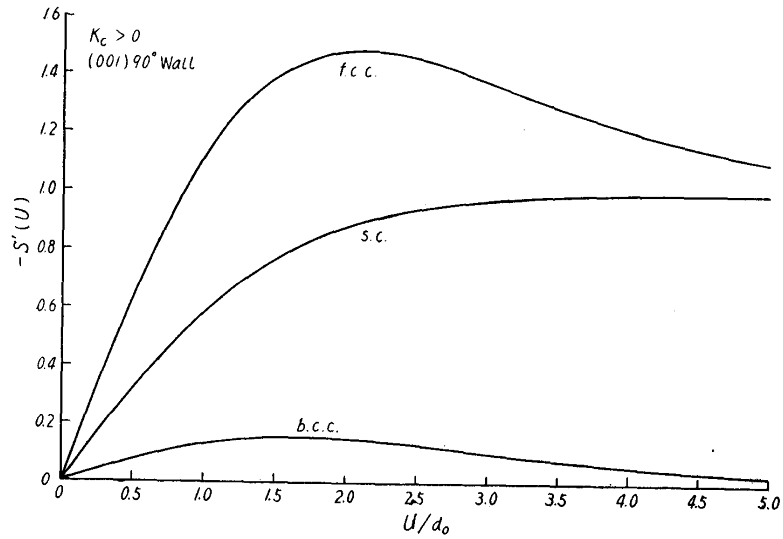

Fig. 1. $-S'(U)$ vs. U/d_0 curves of (100) 90° domain wall for $K_c>0$.

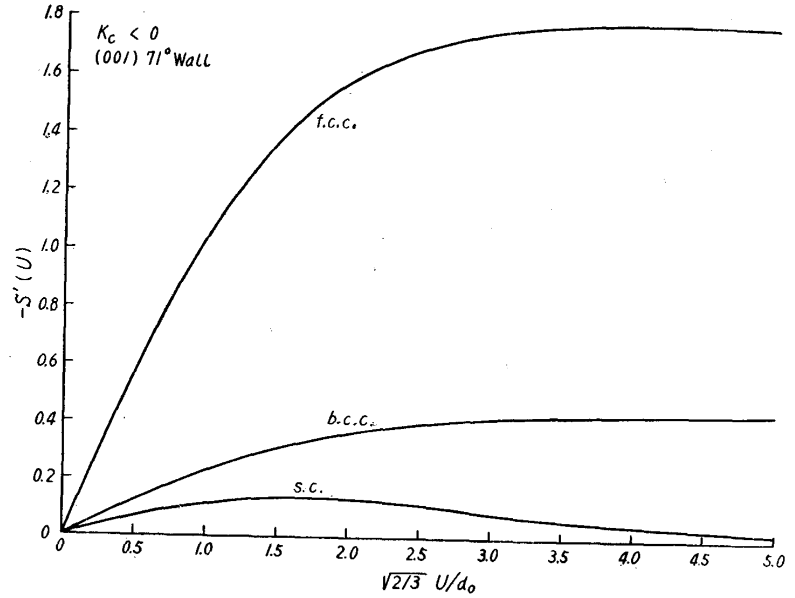

Fig. 2. $-S'(U)$ vs. $\sqrt{2/3}U/d_0$ curves of (100) 71° domain wall for $K_c<0$.

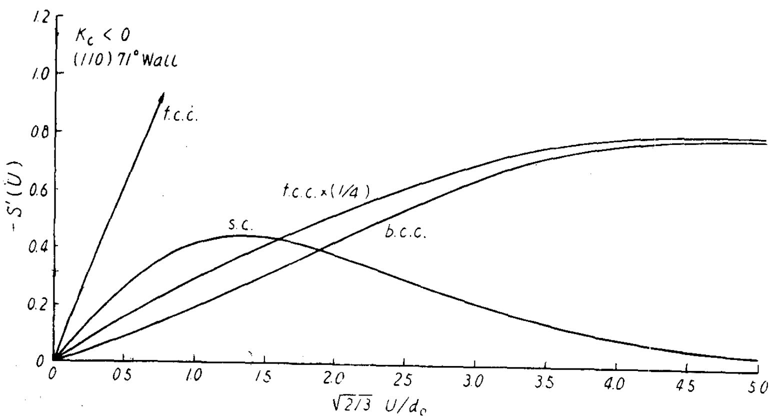

Fig. 3. $-S'(U)$ vs. $\sqrt{2/3}U/d_0$ curves of (110) 71° domain wall for $K_c<0$.

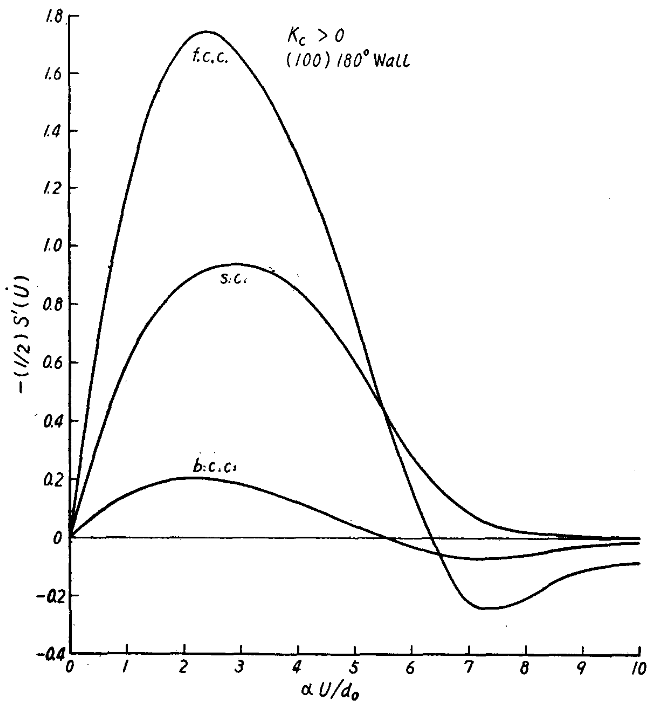

Fig. 4. $-(1/2)S'(U)$ vs. $\alpha U/d_0$ curves of (100) 180° domain wall for $K_c>0$.

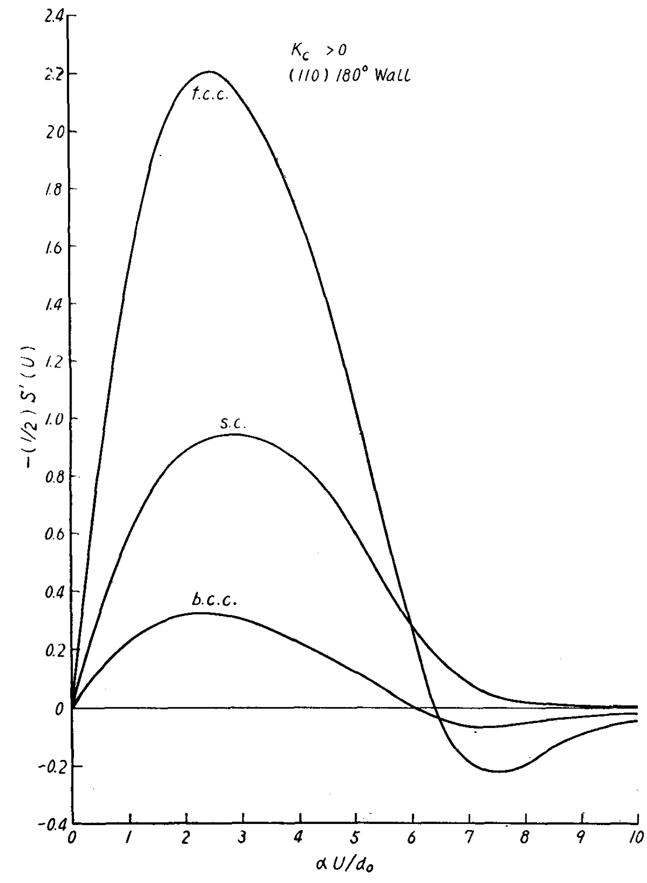

Fig. 5. $-(1/2)S'(U)$ vs. $\alpha U/d_0$ curves of (110) 180° domain wall for $K_c>0$.

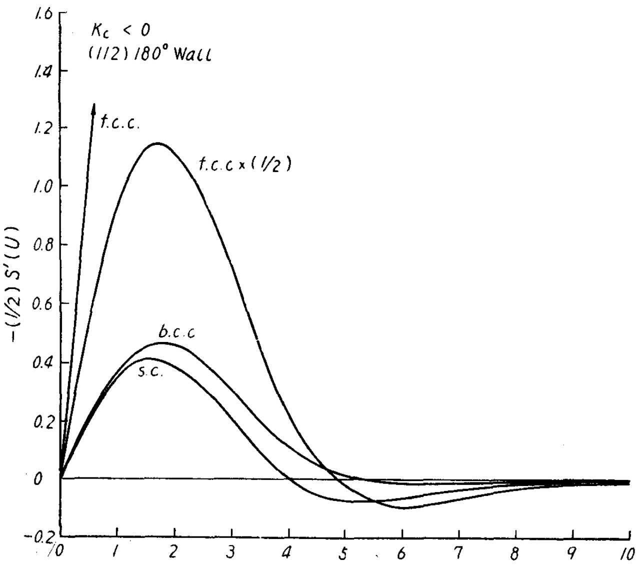

Fig. 6. $-(1/2)S'(U)$ vs. $\alpha U/d_0$ curves of (112) 180° domain wall for $K_c<0$.

that generally the shape of the curve does not depend so much on which plane lies the wall of each type.

Now, let us consider a domain wall under the combined action of the restoring fore, $P(U)$, and of an applied magnetic field, H. As an example, we consider again a (100) 90° wall in face-centered cubic solid solution, separating two domains whose magnetization vectors are directed to the [010] direction in $X>0$ and to the [001] direction in $X<0$. The applied field is supposed to be oriented to the [001] direction, and accordingly the wall may displace towards the [100] direction. Then, if we neglect obstacles for displacement of domain walls other than that due to the stabilization, the displacement, U, below the value corresponding to the maximum of $-S'(U)$ is reversible and is determined by a relation

$$HI_s + P(U) = 0\,, \tag{26}$$

where I_s is the spontaneous magnetization, and the magnetization in the [001] direction induced by this wall displacement, I, is given by

$$I = I_s UA\,, \tag{27}$$

where A is the area of the wall in unit volume. Accordingly, the susceptibility χ for H is given, from Eqs. (26), (27) and (11), by

$$\chi = I/H = -I_s^2 AU/P(U) = -I_s^2 AU/K_u'S'(U)\,. \tag{28}$$

As seen from Fig. 1, for small U, $-S'(U)$ increases almost linearly with U and accordingly the susceptibility χ is practically independent of U and hence of H. After the wall has displaced over the maximum of $-S'(U)$, it displaces irreversibly when the applied field is increased further. If the field is decreased from any position of the curve, the wall is pulled back to the original position by the restoring force equivalent to the reverse field $H_r = P(U)/I_s$ and when the field is reduced to zero the wall comes back to its original position.

For 180° walls, the displacement U below the value corresponding to the maximum of $-S'(U)$ is also reversible and approximately proportional to H. But, after the wall has once displaced irreversibly over the maximum of $-S'(U)$, the wall can not always come back to its original position even if the field is reduced to zero, since the restoring force becomes zero as $U\to\infty$, by circumstances after changing its sign to positive.

Magnetization and hysteresis curves due to the displacement of a domain wall with a $-S'(U)$ curve are schematically shown in Figs. 7(a)~(d). It is to be noted that, even if there are not any obstacle for displacement of domain wall other than that due to the effect of stabilization, there can exist a critical field for the irreversible domain wall displacement or for a conspicuously rapid increase in magnetization, below which the magnetization increases very slowly and quite reversibly, and that the shape of hysteresis curve due to the irreversible domain wall displacement can be very peculiar as compared with those of ordinary ferromagnetics.

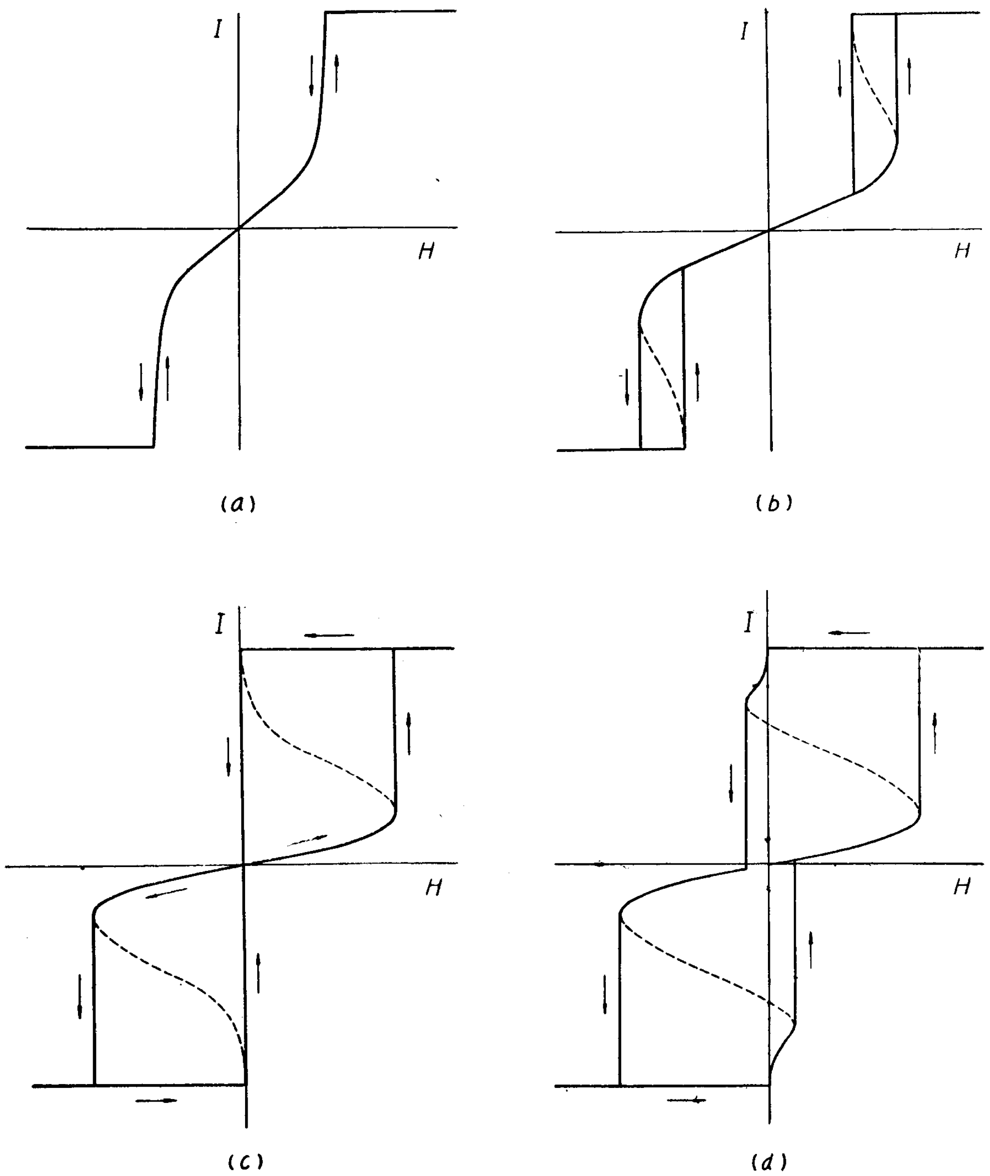

Fig. 7. Schematic magnetization curves due to the displacement of domain walls with $-S'(U)$ curves subscribed, for example, by (a) s.c. in Fig. 1, (b) f.c.c. in Fig. 1, (c) s.c. in Fig. 4, or (d) f.c.c. in Fig. 4. The $-S'(U)$ curve for each magnetization curve is shown by broken line.

So far we have considered the case where the anisotropic distribution of solute atoms does not change during measurement. At high temperatures, however, the diffusion of atoms may proceed with a considerable speed and these solute atoms may redistribute in compliance with the new positions of domain walls established by an applied field. Then, $P(U)$ decreases with a relaxation time determined by the diffusion velocity, so that the magnetic aftereffect may occur.

IV. The perminvar problem

It has been well known that some annealed iron-nickel-cobalt alloys, particularly in baked state, show interesting magnetic behaviors such as the conspicuously constant permeability and low hysteresis loss in low fields and peculiarly-shaped hysteresis loop in higher fields. These alloys are called "perminvar"(4) and the typical composition is 25 percent cobalt, 45 percent nickel and the rest iron (45–25 perminvar). Some investigators(9,10,11) seeked to interpret the origin of these characteristic properties. Iida(11) derived a hysteresis loop similar to that of perminvar, using a model of linear assemblage of 90° domains stabilized by a uniaxial anisotropy and assuming that the magnetization is induced by the rotation of magnetization vectors against the uniaxial anisotropy alone. His theory, however, seems to be not generally applicable for solid solutions, since it neglects the rôle of the magnetocrystalline anisotropy in the magnetization process and assumes that the origin of the uniaxial anisotropy is the heterogeneous formation of superlattice which happened to exist in iron-nickel alloys. It is to be noted, further, that Iida's theory gives the remanence far greater than that obtained experimentally and that it can not refer to the conspicuously constant permeability and low hysteresis loss in low fields.

Now, in iron-nickel-cobalt alloys, the perminvar-type magnetic properties can be found in almost all of face-centered cubic alloys with relatively high Curie temperatures and in body-centered cubic alloys with negative cubic anisotropy constants(4). The consideration described in Section III indicates that the constant permeability and low hysteresis loss in low fields are properties common to *these* cubic solid solution alloys in slowly cooled or baked state(12). However, the following two points might be doubtful :- (1) Does our theory explain the constancy of permeability continuing up to a magnetic field of one oersted or more, as the experimental results(4) indicates? and (2) if it were the case, does a domain wall

(9) O. van Auwers and H. Kühlwein, Ann. Physik, [5] **17** (1933), 121.
(10) S. Kaya and M. Nakayama, Z. Phys., **112** (1939), 420.
(11) S. Iida, cited by S. Kaya, Rev. Mod. Phys., **25** (1953), 49.
(12) When the measurement is done at relatively low temperatures, including room temperature, where the diffusion of atoms can not occur practically, the results of considerations in Sec. III do not strictly hold, since they are obtained under the condition that the domain wall is displaced at a temperature where it is stabilized. But, since the change in the width of domain wall may be negligibly small at low temperatures in solid solutions with relatively high Curie temperatures, we may use, here, the results obtained in Sec. III with sufficient accuracy if we take as K_u' in the equations in Sec. III the value of K_u in Eq. (2), where Θ, in this case, means a equilibrium temperature determined by the conditions of heat-treatment.

really displace from its original position only by a distance as short as its width in such a field range?

For the first question, we consider the 90° wall described in Section III. This is the most favorable case for the displacement of domain wall, since the direction of applied magnetic field coincides with that of the magnetization vector in one domain. The order of magnitude of the maximum field for the constant permeability, H_m, may be given, from Eqs. (26) and (11), by

$$H_m \approx K_u'/I_s \qquad (29)$$

since $-S'(U) \approx 1$ as may by seen from Fig. 1. Taking $I_s = 10^3$ and $K_u' = 10^3$ erg/cm^3, H_m becomes one oersted which is comparable with the experimental results on baked 45–25 perminvar(4).

For the second question, we calculate the total area of domain wall, A, necessary to yield the initial susceptibility of a reasonable order of magnitude. From Eq. (28), the initial susceptibility of polycrystals, $\bar{\chi}_0$, may be given approximately by(13)

$$\bar{\chi}_0 \approx d_0 A I_s^2 / K_u', \qquad (30)$$

since, from Fig. 1, $-U/S'(U) \approx d_0$ for small U, and taking $I_s = 10^3$ and $K_u' = 10^3$ erg/cm^3 as before and $\bar{\chi}_0 = 20$(4), $d_0 A$ becomes about 2×10^{-2}cm^3. This value is somewhat larger in the order of magnitude than that obtained by Néel(1) for iron in his theory of magnetic aftereffect. But, our(14) recent experiments on the effect of heat-treatment on the domain structure of 40 percent cobalt-nickel single crystal have indicated that the domain size in annealed state is far smaller than that in quenched state or that of pure metal, indicating that the domain size can also be influenced by the appearence of the uniaxial anisotropy. Taking into considerations of this fact, the above mentioned value for $d_0 A$ seems reasonable for annealed perminvars. This fact seems to show, in turn, that the permeability in annealed state is reduced appreciably by the effect of stabilization as compared with that in quenched state where the stabilization can not occur.

As seen from Figs. 1~3, every non−180° wall in ferromagnetic cubic solid solutions intends to return back to the original position, even if it has displaced far away, since it is pulled back by the restoring force equivalent to a reverse field of about $K_u'/I_s \approx 1$ Oe. On the contrary, 180° walls have not such a tendency when they have displaced over the maximum of their $-S'(U)$ *vs.* U relations (Figs. 4~6). But, in such a complicated domain structure as observed actually in annealed solid solutions showing the perminvar-type magnetic behaviour(14), every 180° wall is connected to non−180° walls so tightly and so complicatedly that it may return back to almost the same position as the original one. It may be expected, therefore, that the domain distribution hardly changes by demagnetization along any crystal direction and this has been confirmed experimentally(14). These

(13) Since the initial susceptibility is reduced appreciably by the effect of stabilization in solid solutions concerned here, the obstacles for the displacement of domain walls other than that due to the effect of stabilization can be neglected.

(14) M. Yamamoto, S. Taniguchi and K. Aoyagi, Phys. Rev., in the press.

may be the reasons why the remanence is very low as compared with that of pure metals and the descending branch of hysteresis loop often shows a convex towards the abscissa in low fields (cf. Fig. 7)

It has been found that the constancy of permeability in low fields of perminvar is destroyed appreciably by being magnetized in field higher than H_m, and accordingly also by alternating-field demagnetization, commonly employed and that, in order to recover it, it is necessary to bake perminvar once more at a moderate temperature, usually at 425°C[(4)]. This can be interpreted as follows :- As indicated above, domain walls can return back to almost, but not exactly, the same positions as the original ones and these slight deviations from the original positions affect the permeability appreciably, since the constancy of permeability arises from the wall displacements of the order of the width of domain walls from the originally stabilized positions and the magnitude of permeability is reduced appreciably by the stabilization, especially in this range of wall displacement. Then, to recover the constancy of permeability, the uniaxial anisotropy appropriate to new positions of walls must be induced. As shown above, the larger is the uniaxial anisotropy constant, the greater is the restoring force. Accordingly, the constancy of permeability becomes conspicuous by baking at a temperature as low and for a time as long as possible. Usually 425°C is employed as the baking temperature for perminvars, since at a temperature lower than it, the establishment of the equilibrium state may require a very long time, the rate-controlling factor being the relaxation time for an anisotropic distribution of atom pairs by diffusion of atoms, and moreover, the domain structure may not vary appreciably during cooling down from 425°C to room temperature[(12)].

When perminvar is brought to higher temperatures, the maximum field, H_m, below which the permeability can be regarded practically as constant, decreases gradually while the permeability increases[(4)]. These variations may be due to the fact that, according to our theory, K_u' in Eq. (29)[(12)] decreases in proportion to B_T^2, or approximately to j_T^2, where j_T means the ratio of spontaneous magnetization at a temperature T and that at absolute zero. At very high temperatures, the diffusion of atoms becomes so rapid that the anisotropic distribution of atom pairs, which restores domain walls, may change during measurement and, accordingly, the effect of stabilization can not be detected by statical measurements. Dynamical measurement, however, may detect the effect as the magnetic aftereffect or relaxation in magnetization at these temperatures.

If an external tension is applied, the hysteresis loop of 35–30 perminvar looses its peculiar shape gradually and finally becomes normal at about 6.8 kg/mm^2[(4)]. This may be due to the fact that the stabilization effect is exceeded by the magnetoelastic energy associated with the applied stress. Then, the minimum magnetoelastic energy responsible for such a change in the shape of hysteresis loop may be roughly equal to the uniaxial anisotrypy induced. Thus, the uniaxial anisotropy induced in 35–30 perminvar is estimated to be about 700 erg/cm^3, taking $\lambda = 12\times10^{-6}$[(15)],

(15) See, for example, R.M. Bozorth, *loc. cit.*, p. 674.

which is a reasonable value.

In conclusion, we may say that the perminvar-type magnetic properties arise from the displacement of domain walls stabilized by the uniaxial anisotropy induced in cubic ferromagnetic solid solutions and they are the most common properties of solid solutions in which the uniaxial anisotropy can be induced along the directions of easy magnetization. Yamamoto and the present author(16), indeed, observed the perminvar-type magnetic properties in annealed state of face-centered cubic solid solutions in binary nickel-cobalt alloy system, in which any phase change such as the superlattice formation has not yet been found.

V. The influence of heat-treatment on the magnetic properties of ferromagnetic cubic solid solutions

In the preceeding section, we have shown that the magnetic properties of perminvars are really not peculiar but common to ferromagnetic cubic solid solutions in which the uniaxial anisotropy can be induced along the directions of easy magnetization. When such a solid solution is quenched from above the Curie temperature, the stabilization of domain walls can not occur and accordingly the magnetic properties in quenched state may be different from those in annealed state. We shall exhibit the general aspect of the effects of various heat-treatments on the magnetic properties of solid solutions in the following. It is hardly needless to note, however, that the magnetic properties may also be affected by imperfections in the specimen as well as by the mutual interaction of domains so sensitively and so complicatedly that we can not expect to find a complete and quantitative agreement between theory and experiment.

A. Face-centered cubic solid solutions and body-centered cubic solid solutions with negative cubic anisotropy constants, namely solid solutions in which the uniaxial anisotropy can be induced along the directions of easy magnetization

The magnetic properties of these solid solutions in quenched state where the domain walls are not stabilized are determined in the same way asthose in pure metals in annealed state, only if the thermal stress due to quenching were negligible. Then, their magnetic properties can be compared with those of pure metals only in quenched state, which may be called the standard state of solid solutions.

In baked or slowly cooled state, however, every domain wall is stabilized and thus the perminvar-type magnetic properties appear, as fully discussed in the preceeding sections. Accordingly, the magnetization curve in annealed state is less steep than that in quenched state. Moreover, the domain structure is very complicated and small, and the domain distribution is hardly changed by alternating-field demagnetization, maintaining an almost isotropic distribution among all of the directions of easy magnetization. It may be expected that the displacement

(16) M. Yamamoto, S. Taniguchi and K. Hoshi, Nippon Kinzoku Gakkai-shi, **17** (1953), 615; Sci. Rep. RITU, **A6** (1954), 539; M. Yamamoto and S. Taniguchi, Nippon Kinzoku Gakkai-shi, **19** (1955), 645 and 651.

of non -180° walls contribute appreciably to the magnetization process.

Finally, when such solid solutions are annealed with the presence of an externally applied magnetic field, which is strong enough to saturate the specimen, the uniaxial anisotropy is induced along the direction of magnetization or of the magnetic field applied during annealing. Then, the direction of easy magnetization are determined by the combined effect of the cubic and the uniaxial anisotropies and the cubic solid solutions behave as magnetically uniaxial, namely, the whole crystals in the specimen are divided almost entirely into 180° domains and the magnetization proceeds mainly by the displacement of 180° walls. Moreover, every domain wall is not stabilized and further the displacements of domain walls are easier than in quenched state, since the restraining force due to imperfections of some types may be lessened by the appearance of homogeneous uniaxial anisotropy. The remanence becomes greater than that in the case of annealing without magnetic field and, as the cubic anisotropy decreases, it approaches to the saturation magnetization.

As an actual example, the magnetization curves of a polycrystalline specimen of 40 percent cobalt-nickel alloy subjected to the above-mentioned three kinds of heat-treatment are shown in Fig. 8[16,17]. It is to be noted that, according to our

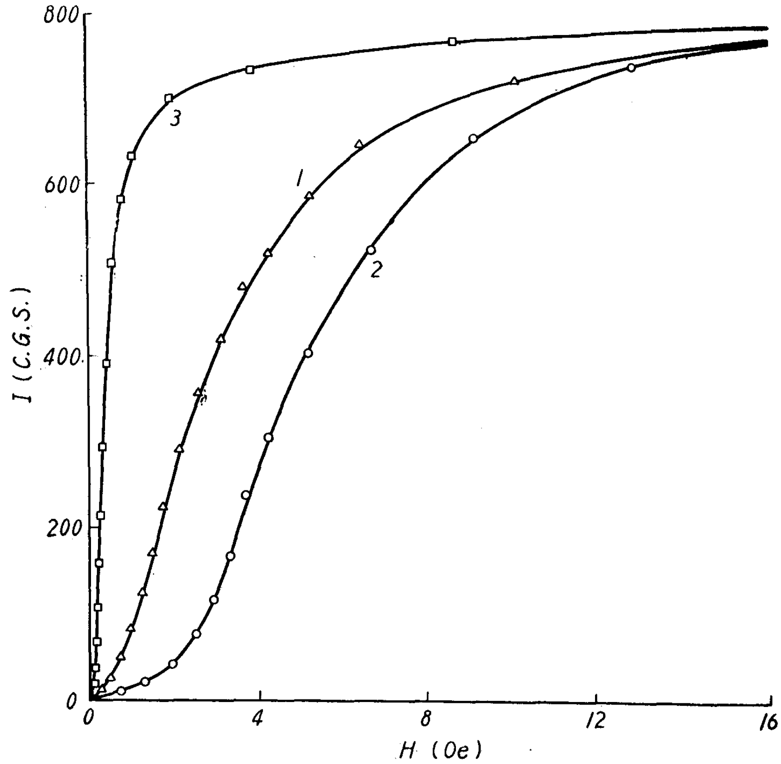

Fig. 8. Effect of various heat-treatments on the magnetization curve of 40.6 percent cobalt-nickel alloy[16,17].
1 : air-quenched from 750°C, 2 : annealed without magnetic field, 3 : annealed with magnetic field.

(17) M. Yamamoto, Nippon Kinzoku Gakkai-shi, **11** (1947), No. 11–12 (P. 3) ; **13** (1949), No. 6 (P. 15) ; Sci. Rep. RITU, **A4** (1952), 14.

theory, the difference between curves (1) and (3) must be smaller than that between (1) and (2), in contradiction to the experimental results, since the absolute value of the cubic anisotropy is fairly large in this alloy (5.4×10^4 erg/cm^3)[17]. This disagreement may be due to the fact that quenching from 750°C to copper plate does not completely satisfy the quenching condition mentioned above.

B. Body-centered cubic solid solutions with positive cubic anisotropy constants

Since the uniaxial anisotropy cannot be induced along the directions of easy magnetization in these solid solutions and accordingly the effect of domain wall stabilization is very small, the perminvar-type magnetic properties hardly appear even in baked state. Then the difference between magnetization curves in quenched and in baked states is very small as compared with the former case, though the difference between those in magnetically annealed and in quenched or baked states may be almost the same as in the former case.

As an example of the difference in behavior between magnetization curves of body-centered cubic solid solutions with positive and negative cubic anisotropy constants, the magnetization curves of 30 and 75 percent cobalt-iron alloys are shown in Fig. 9[18]. It has been well known that, in iron-cobalt alloys, the superlattice FeCo is present and the magnetostriction increases appreciably with ordering. Accordingly, the variation in magnetization curve due to the change in heat-treatment can not be attributed to the appearance of the uniaxial anisotropy alone.

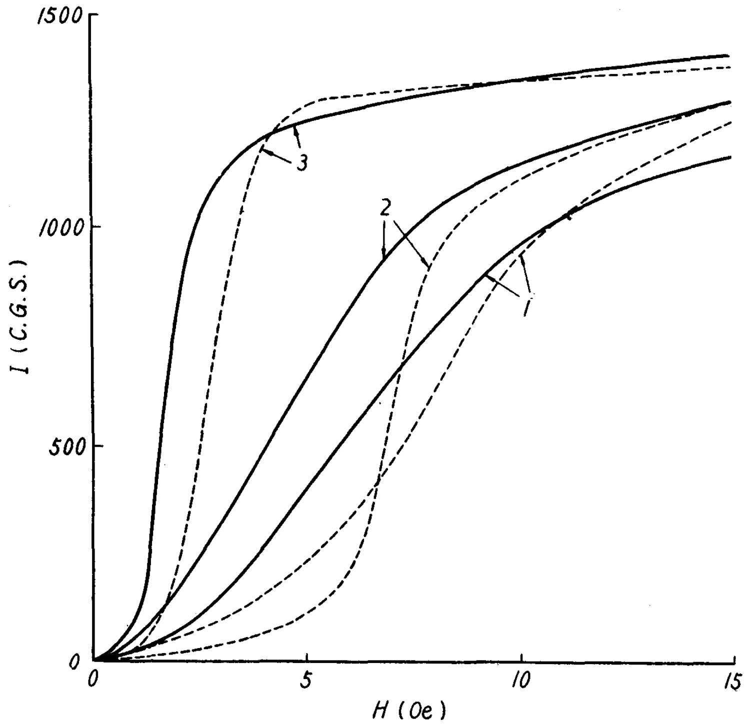

Fig. 9. Effect of various heat-treatments on the magnetization curves of 30 and 75 percent cobalt-iron alloys[18].
—— 30%Co ········ 75%Co
1 : furnace-cooled, 2 : baked, 3 : annealed with magnetic field.

(18) S Kaya and H. Sato, Proc. Phys.-Math. Soc. Japan, **25** (1943), 261.

However, the compositions of the alloys concerned are distant from the stoichiometric composition of superlattice by a considerable and nearly the same amount, so that the relative difference in behavior of the two alloys may be referred to here. As seen from Fig. 9, the magnetization curves of the two alloys in magnetically annealed state (3) lie above those in furnace-cooled state (1), but, in low fields, the magnetization curves in baked state (2) of 30 percent coblat alloy lies above, while that of 75 percent cobalt alloy lies below, the curves in furnace-cooled state (1). Moreover, only the magnetization of baked 75 percent cobalt alloy increase first linearly up to about 4 oersteds, indicating the constant permeability. These different behavior may be comprehended by the fact that the sign of cubic anisotropy constant is positive in iron-cobalt alloys containing less than about 46 percent coblat, and becomes negative beyond this composition(19). This may also be the reason why only binary iron-cobalt alloys containing more than about 50 percent cobalt belong to the perminvar region(4).

The effect of magnetic annealing has been estimated generally in comparing the magnetic properties in magnetically annealed state with those in annealed state. Then the appreciable effect may be expected, in general, for face-centered cubic solid solutions and the body-centered cubic solid solutions with negative cubic anisotropy constants as well as for solid solutions with very small cubic anisotropy constants, while the appreciable effect of quenching may be expected only for the former two solid solutions.

Finally, we shall refer briefly to the *permalloy problem* (20,21), which is to explain the characteristic properties posessed by permalloys (face-centered cubic nickel-iron alloys). The magnetic characteristics of permalloys are expressed usually as(21) :- "They have unusually high permeabilities which are maximum at about 79 percent nickel, the permeabilities are influenced markedly by heat-treatment and specifically are increased by cooling rapidly, and a magnetic anneal causes drastic change in various magnetic properties." In the present author's opinion, however, it seems reasonable to rewrite the above expression as follows :- (1) They have unusually high permeabilities which are maximum at about 79 percent nickel *in rapidly cooled state*, (2) *their permeabilities are reduced appreciably by slow cooling and a minimum in permeability occurs at about 65 percent nickel*, and (3) a magnetic anneal causes drastic change in various magnetic properties. These characteristic properties seem to be caused by three factors. The first of them is the concentration dependence of the material constants affecting the magnetic properties, the second their changes with the formation of superlattice Ni_3Fe, and the third the appearance of the uniaxial anisotropy. The first characteristic of permalloys (1) may be interpreted approximately by the first factor, as has been emphasized by Bozorth(21). The second characteristic (2), however, can not be interpreted by the second factor alone, since, for example, the permeability of

(19) J. W. Shih, Phys. Rev., **46** (1934), 139.
(20) See, for example, R.M. Bozorth, *loc. cit.* P. 102.
(21) R. M. Bozorth, Rev. Mod. Phys., **25** (1953), 42.

65 percent nickel alloy decreases appreciably by slow cooling, but both the cubic anisotropy constant and magnetostriction constant in the direction of easy magnetization also decreases considerably by the same heat-treatment. Moreover, this alloy in slowly cooled state shows a hysteresis loop of the perminvar type[20]. Accordingly, the second characteristic must be interpreted mainly by the third factor, that is, by the domain wall stabilization discussed above, though it may be partly due to the second factor, particularly in a composition range near 75 percent nickel[22]. The third characteristic (3) is not characteristic to nickel-iron alloys but quite common to cubic solid solutions, as indicated above, though the drastic effect in permalloys may be resulted from the first factor, particularly from the fact that the cubic anisotropy happens to be small in all the composition range concerned. Since the uniaxial anisotropy induced by magnetic annealing is larger, the higher are the concentration of solute atoms and the Curic temperarure, the most pronounced effect in magnetic anneal may be expected to occur at about 65 percent nickel as in the cases of the peculiar shape of hysteresis loop and of the lowering of permeability in slowly cooled state.

Summary

Since, as indicated in the previous papers[2], the uniaxial anisotropy induced by magnetic annealing in ferromagnetic solid solutions can be interpreted as caused by an anisotropic distribution of solute atom pairs and it depends on the direction of magnetization vector relative to the crystal axes, the uniaxial anisotropy may be induced in compliance with the distribution of magnetization vectors corresponding to a domain structure present during heat-treatment. As a result, a domain wall is acted by a large restoring force, which varies with its displacement and corresponds to the magnetic field of one oersted or more. In other words, the domain wall is stabilized by the induced uniaxial anisotropy and the permeability in low fields is reduced appreciably by the effect of stabilization. The calculated functional form of this restoring force leads to a conspicuous constancy of permeability and reversible displacement of domain walls in low fields and to the low remanence for face-centered cubic solid solutions and body-centered cubic solid solutions with negative cubic anisotropy constant, in which the uniaxial anisotropy can be induced along the directions of easy magnetization. Thus, the peculiar magnitic properties such as possessed by perminvars have been shown to be common to these solid solutions. Moreover, it has been shown that available experimental results on perminvars, as to the change in the constancy of permeability by demagnetization with alternating field, magnetic behavior in high temperature, effect of applied tension on the shape of hysteresis loop, etc., can be interpreted in terms of the idea of the stabilization of domain walls by the induced uniaxial anisotropy.

Further, the influence of heat-treatments on the magnetic properties of cubic

(22) In supermalloys, where the highest permeabilities are attained by slow cooling, the stabilization of domain walls may be suppressed by lowering the Curie temperature below 400°C. We shall discuss on supermalloys in a later paper.

solid solutions have been discussed and it has been emphasized that the standard state which is comparable with pure metals is the state quenched from above the Curie temperature, in which the domain wall is not stabilized by the induced uniaxial anisotropy. The difference in magnetic behavior of binary iron-cobalt alloys containing more than and less than about 50 percent cobalt has been interpreted by the fact that the uniaxial anisotropy can not be induced along the [100] directions, which is the directions of easy magnetization for ferromagnetic crystals with positive cubic anisotropy constants. Finally, it has been pointed out that the permalloy problem may also be clarified by introducing the effect of domain wall stabilization.

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